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## **Parameters Influencing Emissions of PCDDs/Fs from Open Burning of Household Waste in Barrels**

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### **ABSTRACT**

The uncontrolled burning of household waste in barrels has recently been implicated as possibly being a major source of polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDDs/Fs). Previous measurements of PCDDs/Fs from controlled studies of burn barrels (holding bulk density constant) resulted in a very high degree of variability, even between nominally identical test conditions. It is hypothesized that slight differences in waste composition, distribution of various waste components in the barrel, and bulk waste density could significantly influence emissions of PCDDs/Fs from this combustion source. This paper will report on the results from tests that were specifically designed to examine some of the factors influencing the emissions of PCDDs/Fs from burn barrels. Based on the wide variability of emissions within duplicate run sets in spite of all efforts to replicate run conditions, and the large number of interacting variables that can account for this variability, it is suggested that a probability-based scheme using Monte Carlo simulations may be useful for developing emissions inventory estimates for this source.

### **INTRODUCTION**

#### **Previous Results**

The uncontrolled burning of household waste in barrels is commonly practiced in rural areas of the U.S. and is a primary waste management technique in many parts of the developing world. This activity typically consists of a household's placing their garbage in a 208 L (55 gal.) drum called a “burn barrel” and, when a sufficient quantity of

garbage has accumulated in the burn barrel, igniting the waste, and burning it over a period of time lasting several hours.

The EPA's Control Technology Center (CTC) received numerous requests from state and local agencies requesting information on emissions from burn barrels. In response to these requests, the CTC funded a study to characterize the emissions of many pollutants from a limited number of burn barrels. This study was performed by EPA's Air Pollution Prevention and Control Division (APPCD), in collaboration with the New York State Department of Health (NYSDOH). Pollutants measured during this study included fixed combustion gases (e.g., carbon monoxide [CO] and nitrogen oxides [NO<sub>x</sub>]), volatile organic compounds (VOCs), semivolatile organic compounds (SVOCs), particulate matter (PM), polychlorinated biphenyls (PCBs), and polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDDs/Fs). The results from this study were published in an EPA report in 1997 [1]. Emissions of many pollutants were reported to be significant. Emissions of PCDDs/Fs were significant enough that burn barrels could potentially be a major national source of airborne PCDDs/Fs, given moderate estimates of frequency of this practice. In addition, the 1997 report showed significant variability between duplicate runs.

Lemieux et al. performed further data analyses on the PCDD/F data from the 1997 report, in an attempt to see if any trends could be observed that may suggest a causal relationship between waste composition, burn conditions, and emissions of PCDDs/Fs [2]. Although apparent relationships between PCDD/F emissions and airborne hydrochloric acid (HCl) and copper (Cu) emissions were observed, sufficient data were not available to conduct rigorous statistical analyses.

The limited amount of data and high degree of variability confounded efforts to incorporate burn barrels into the U.S. dioxin inventory [3]. In an effort to reduce the uncertainty in the emissions estimation, EPA's Office of Pesticides, Prevention, and Toxic Substances (OPPTS) funded additional testing on burn barrels so that the emissions of PCDDs/Fs could be characterized as a function of waste composition, burn conditions, and other physical properties of the waste in the barrels (e.g., degree of compaction and wetness). Initial results from these follow-up tests were reported by Gullett et al. [4,5], where it was found that waste composition parameters (e.g., chlorine content, Cu emissions) and combustion conditions (e.g., barrel temperature distributions, and CO concentrations) can statistically account for the variability between runs.

## **PCDD/F Formation**

Extensive research has been conducted to investigate the formation mechanisms of PCDDs/Fs in the years since PCDDs/Fs were first discovered in the exhaust gases from municipal waste combustors (MWCs) in 1977 [6]. Since then, multiple formation mechanisms have been proposed [7-13]. Field studies on MWCs have shown that the amount of flyash (and its accompanying metallic catalysts) and organic precursors that pass through the temperature window, between 250 and 700 °C, as well as the amount of time spent in that optimal temperature window are the primary variables affecting PCDD/PCDF emissions [14]. Field studies have been unable to clearly demonstrate a

correlation between chlorine (Cl) input and PCDD/F emissions in full-scale combustion systems, probably because other variables (PM carryover, PM control device temperature, and combustion efficiency) dominate [15]. It is the authors' view that Cl is present in excess relative to the other reactants in these systems. Based on the current PCDD/PCDF formation theories, a "worst-case scenario" for formation of PCDDs/Fs from combustion systems would be a combination of the following features:

- poor gas-phase mixing,
- low combustion temperatures,
- oxygen-starved conditions,
- high PM loading,
- PM-bound copper,
- presence of HCl and/or Cl, and
- significant gas-phase residence time in the 250-700 °C temperature range.

Barrel burning is a combustion source that appears to fit all of the requirements for such a worst-case scenario for production of PCDDs/Fs.

This paper focuses on an analysis of the variability of the PCDD/F data in identical runs.

## **EXPERIMENTAL**

Combustion studies were performed at the EPA's Open Burning Test Facility or "burn hut" [1] to provide an initial determination of the impact of limited variation in waste composition on combustion conditions and PCDD/F emissions from a simulated domestic, backyard barrel burn. A composition representative of domestic household waste was prepared for testing based on the typical percentages of various waste materials characterized and quantified by the New York Department of Environmental Conservation's Division of Solid Waste [1]. This synthesized domestic household waste primarily consisted of actual unshredded household waste collected at home by various ARCADIS staff members. Each batch was constructed of the same specific waste types combined together such that each test had the same composition with the exceptions noted in Table 1. Each test consisted of 6.8 kg (15 lb) of waste, randomly mixed for a brief time in a concrete mixer and dumped en masse into the test container. Variation from the baseline composition (0.2 % by weight polyvinyl chloride, PVC) consisted of testing at three different PVC levels (0.0, 1.0, and 7.5 % by wt) using pipe forms. PVC levels were effected through substitution of high density polyethylene (HDPE) and iron conduit (both also in pipe form), in an effort to approximate consistent physical and energy properties of the waste across all batches while varying Cl. Inorganic Cl levels were derived by soaking the paper portion of the waste in a calcium chloride ( $\text{CaCl}_2$ )-based deicer followed by drying. Additional plastic components included polystyrene (PS) and polyethylene terephthalate (PET).

To represent the most common practice for residential waste burning, the test container consisted of an aged, 208 L (55 gal) steel barrel with twelve 2 cm diameter ventilation holes around the base. Prior to the testing described in [1], the barrel was sandblasted to remove residual paint and any remaining contents that might affect emissions. The barrel

was placed on an electronic scale platform to allow the mass consumed by combustion to be continuously monitored. An aluminum skirt was placed around the outer circumference of the barrel to minimize the potential for recirculation of combustion gases back through the air inlet holes. High volume air handlers provided metered dilution air into the burn hut. Additional fans were set up inside the burn hut to enhance recirculation within the hut. The hut was lined with Tedlar<sup>®</sup>. Type K thermocouples were inserted into the barrel at regularly spaced heights and radial locations from the bottom, within and above the waste. Before the initiation of each test, the material to be combusted was placed in the barrel, air flow through the facility was initiated, and 10 min of background data were obtained. These data came from continuous emission monitors (CEMs) which sampled for oxygen (O<sub>2</sub>), carbon dioxide (CO<sub>2</sub>), and CO from the gas stream of the exhaust gas duct.

PM with an aerodynamic diameter < 2.5 µm (PM<sub>2.5</sub>) was measured using a dichotomous sampler placed inside the burn hut. Sampling for PCDD/Fs and PCBs (the latter not covered in this paper) was completed via an ambient-air Graseby<sup>™</sup> PS-1 sampler located within the test facility and operated for about 1.5 h at between 0.062 and 0.071 m<sup>3</sup>/min (2.2 and 2.5 ft<sup>3</sup>/min). The PCDD/F/PCB train consisted of an open-faced filter holder followed by a polyurethane foam (PUF)-sandwiched XAD-2<sup>®</sup> bed vapor trap. The combined filter and vapor-phase module was analyzed using high-resolution gas chromatography and high-resolution mass spectrometry (HRGC/HRMS). Sampling and analytical methods followed those reported previously [4].

The material to be combusted was lit for a short period (<3 min) using a propane torch inserted into a hole midway up the side of the barrel. Sampling was initiated at least 2 min after the removal of the propane flame. Samples were collected over the course of the active burn, and sampling was terminated when the burn mass did not change over an extended period. Blank tests (tests without waste combustion) were also sampled to ensure that the sampling and analysis methods, as well as the feed air, were not biasing the test. Estimated emissions of PCDD/Fs per unit mass burned were calculated using the concentration of the pollutant in the sample, the flow rate of dilution air into the burn hut, the run time, and the mass of waste burned. When analyzing and reporting the results, all non-detects (NDs) and incidences of questionable analytes were set to equal zero. International Toxic Equivalency (TEQ) emission values were calculated using toxic equivalent factors (TEFs) from EPA's interim procedures for assessing PCDD/F risk [16].

## RESULTS AND DISCUSSION

Table 2 lists the PCDD/F data from 18 runs designated Runs A through G (runs are listed in chronological order not alphabetical order). The data are represented in nanograms per kilogram waste consumed. Data are also presented in I-TEQ units of nanograms per kilogram waste consumed. The PCDD/F emission values range over many orders of magnitude. Also noted are the data for average and maximum CO concentrations (in parts per million) and the average and maximum temperatures recorded by thermocouple number 6 (TC6) in degrees centigrade. TC6 was mounted just inside the top of the barrel. TC6 values are reported because that thermocouple's measurements were one of

the parameters that showed statistically significant influence on the emissions of PCDDs/Fs during statistical analysis of the data [4,5].

Figure 1 shows a distribution of the homologues from a representative run (Run A). The PCDDs are lower than the PCDFs, and the emissions are less as the number of substituted Cl atoms on the molecule increases. This homologue distribution exhibits both similarities to and differences from those found in MWCs [17]. For example, in the burn barrel samples, the furans were higher than the dioxins, which is an observation that is consistent with MWCs. However, the burn barrel samples showed a decrease in the concentrations of homologues as the number of Cl atoms increases, which is the opposite of the homologue distributions from MWCs. A possible explanation for this observation is that the chlorinated precursors for the PCDDs/Fs were not highly chlorinated, and the resulting condensation products were not highly chlorinated. Previous work [1] showed a marked decrease in the emissions of chlorinated benzenes as the number of chlorines substituted on the benzene ring increased (i.e., monochlorobenzene >> dichlorobenzene >> trichlorobenzene etc).

In an attempt to examine the variability between “identical” runs, results from the five baseline tests (A, B, C, D, and E) were evaluated statistically to see if a normal distribution was present. Concentrations from each measured PCDD/F isomer, as well as the totals from each homologue group, the TEQ, the total PCDD/F, average and maximum CO, and average and maximum TC6, were subjected to a Shapiro-Wilk W test for normal distribution (N=5) using the SAS-JMP software. Table 3 lists the results from the normal distribution test. All of the evaluated parameters displayed normal distributions. In addition, all evaluated PCDD/F-related parameters exhibited standard deviations that were a significant fraction of the mean. For most PCDD/F-related parameters, the difference between the upper and the lower 95% mean values was approximately 1-2 orders of magnitude. Figure 2 is an example distribution curve for the TEQ values. This normal distribution curve is exceedingly flat, with the probability mass function being approximately 0.006 at the mean TEQ value for the baseline tests. This observation suggests that simply using a mean value for calculations of the emission factor may not result in reliable estimates of emissions. Variability in the CO concentrations and temperatures did not exhibit the same degree of variability as the PCDD/F-related parameters.

The likely cause of the high degree of variability between nominally “identical” runs is probably variation in the distribution of the components of the waste from run to run. The waste was composed of discrete items that were mixed together in a cement mixer to randomize their distribution prior to introduction to the barrel. Based on observations of the significant effects of waste composition on the emissions of PCDDs/Fs from burn barrels [4,5], it is apparent that the proximity of certain components of the waste to the flame front and oxygen supply in the barrel could dramatically influence the production rate of PCDDs/Fs in the barrel. At any given time, only a small fraction of the waste in the barrel is combusting. Which waste components are burned together will likely affect PCDD/F formation.

## CONCLUSIONS

A series of tests were performed to investigate emissions of PCDDs/Fs from open burning of household waste in barrels. These tests were systematically performed so that emissions could be characterized as a function of waste composition, physical properties, and combustion-related parameters, and so that variability could be measured between duplicate test conditions.

Five tests were performed at nominally “identical” waste compositions. These tests exhibited significant variations in the emissions of PCDDs/Fs, with a 1-2 order of magnitude spread between the lowest and the highest values for individual isomers, homologue groups, total PCDDs/Fs, and TEQ values. The emissions of all of the measured pollutants and pollutant groups were found to be normally distributed. However, the normal distribution curves that resulted from the statistical analysis were exceedingly flat, showing wide variations between the lowest and highest 95% confidence intervals, and with the standard deviations being a significant fraction of the means.

Thus far, based on the work that has been done, the work that has not been done, and the work that is ongoing, the potentially important parameters that may effect the observed variability in emissions of PCDDs/Fs from barrel burning may include:

Studied variables:

- Cl content
- Inorganic vs. organic Cl
- Bulk density of waste
- Recycler vs. non-recycler

Variables being examined in ongoing work:

- Open pile vs. contained
- Charge size

Variables not yet studied:

- Ambient temperature effects
- Wind speed/number of ventilation holes
- Other compositional factors (e.g., day to day variations, households that compost)
- Ash layer size
- Burn barrel design variants
- Rain or other weather conditions during burn
- Use of accellerant (e.g., gasoline) as ignition source

Since only duplicate tests were performed on other waste compositions and burn conditions, the assumption can be made that the baseline test conditions would reflect process variabilities that are inherent in the burn barrel combustion source. Noting that the emissions from duplicate tests span several orders of magnitude, it can be assumed that similar distributions would be found at other waste compositions and burn conditions. Simply using a mean value may not result in an accurate measure of the

emissions when it comes time to calculate an emission inventory for burn barrels. Rather, a probability-based approach may provide a more reliable measure of the emissions from burn barrels. One such approach could involve combining survey data from the field with a Monte Carlo simulation. The survey data would contain information about how the population actually burn their waste in barrels (e.g., frequency, waste composition, barrel preparation). The Monte Carlo simulation would then use probability theory to analyze many possible cases. Combined, the two techniques would generate an estimate of the contribution of burn barrels to the national dioxin inventory.

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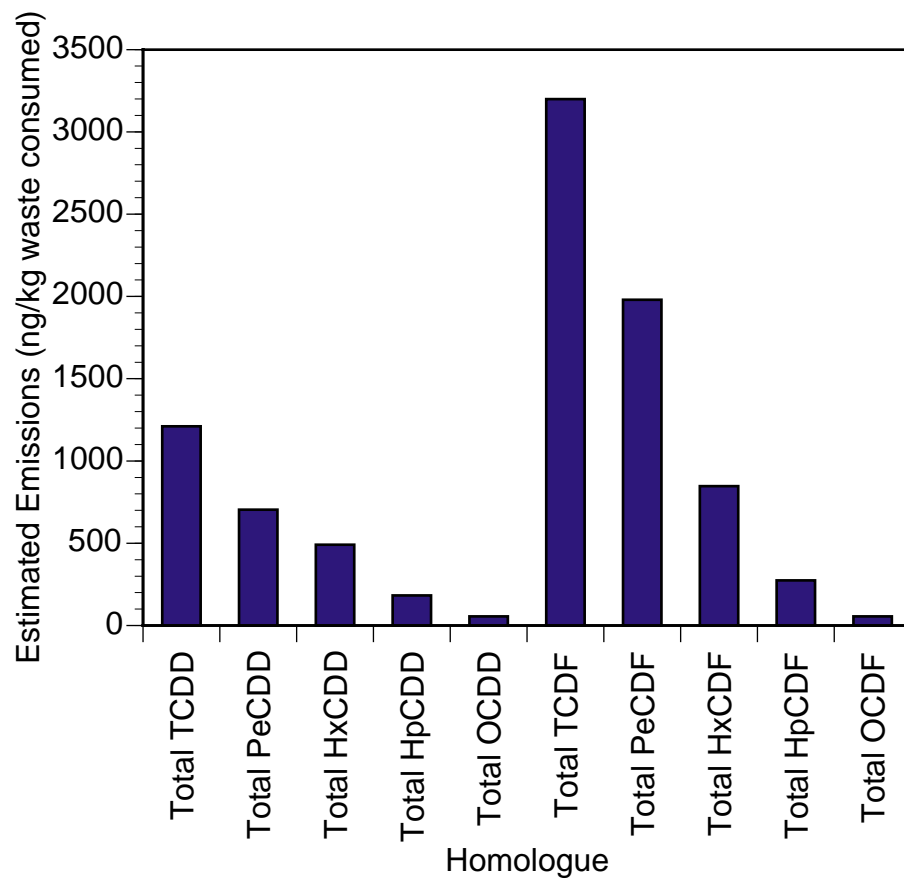


Figure 1. Sample PCDD/F Homologue Profile (Run A).

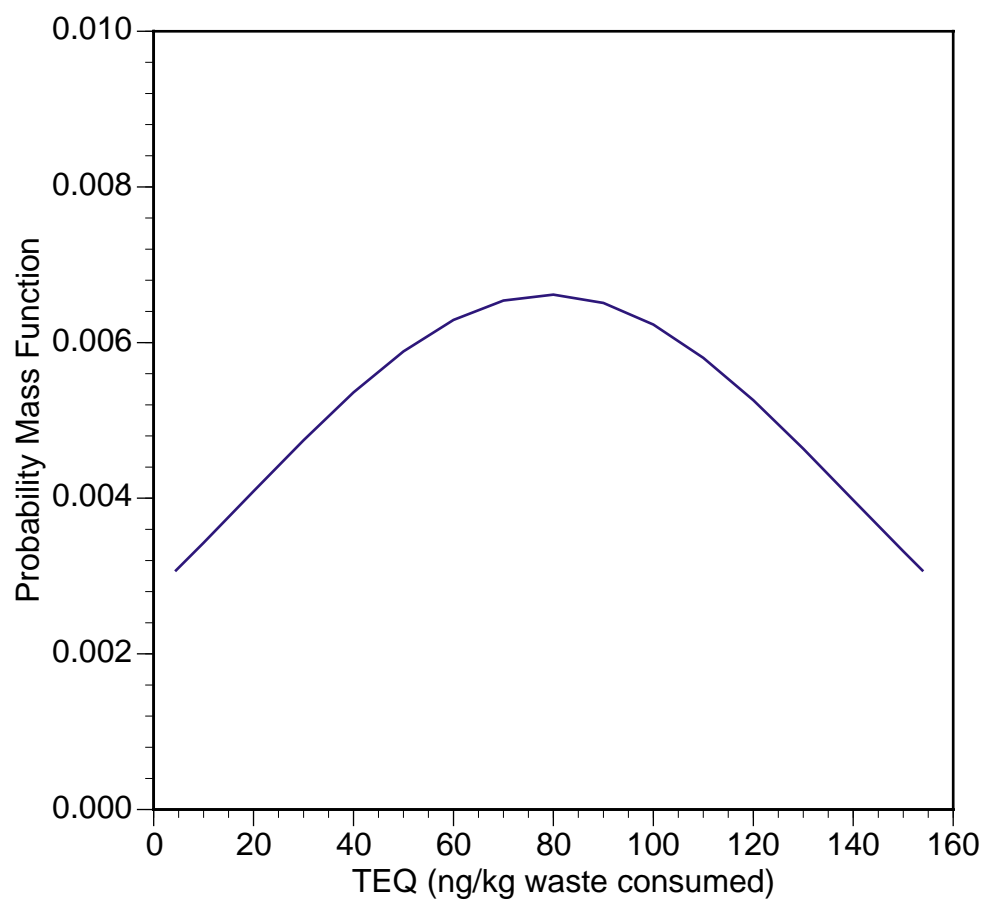


Figure 2. Normal Distribution Function for TEQ.

Table 1. Baseline Waste Composition

Waste Category	Waste Description	Target wt %
Paper	Newspaper, books, office paper	32.8
	Magazines and junk mail	11
	Corrugated cardboard, kraft paper	7.6
	Paperboard, milk cartons, drink boxes	10.3
Plastic Resin	PET #1, soda bottles	0.6
	HDPE #2, detergent bottles, pieces	6.6
	PVC #3, schedule 40 pipe	0.2
	PS #6, food trays	0.1
	Mixed #7, polyester fiber fill	0.1
Food Waste	Frozen processed potatoes	5.7
Textile/Leather	Rubber and leather sneakers	3.7
Wood	Chipboard, plywood	1.1
Glass/Ceramics	Bottles, jars	9.7
	Broken ceramics, flower pots	0.4
Metals (Ferrous)	Iron (cans), dog food cans	7.3
Metals (Non-ferrous)	Aluminum cans, foil, soda cans	1.7
	Wire, copper pipe, batteries	1.1
Total		100

- Inorganic chlorine tests were conducted with calcium chloride ( $\text{CaCl}_2$ ) saturated newspapers (using Prestone Driveway Heat™) such that  $[\text{Cl}] = 7.5 \text{ wt } \%$ . HDPE #2 = 3.3 wt %, 224.53 g; PVC = 0 wt %, 0 g; iron cans = 3.3 wt %, 224.53 g.
- The 0 wt % PVC test consisted of HDPE #2 = 6.7 wt %, 455.86 g; PVC = 0 wt %, iron cans = 7.4 wt %, 503.49 g.
- The high Cu mix test consisted of bottles/jars = 8.7 wt %, 591.94 g; iron cans = 6.4 wt %, 435.45 g; wire, copper pipe, batteries = 3 wt %, 204.12 g.

Table 2. PCDD/F Data (ng/kg waste consumed)

	Test A Baseline	Test B Baseline	Test C Baseline	Test K	Test D Baseline	Test L	Test M	Test O	Test P	Test E Baseline	Hut Blank	Test S	Test T	Test U	Test Q	Test W	Hut Blank	Test G
<b>PCDDs/Fs</b>																		
Total TCDD	1210	130	93	21	24	408	979	4645	12714	606	NA	60	3975	3832	22570	492	NA	672
Total PeCDD	704	98	137	3	4	272	593	4625	12183	463	NA	39	2767	2607	17266	437	NA	369
Total HxCDD	491	123	33	3	4	205	492	5039	14035	456	NA	37	2061	2275	16020	368	NA	478
Total HpCDD	183	83	30	5	6	85	256	3166	9164	225	NA	19	1047	1063	7298	193	NA	253
Total OCDD	56	48	12	11	7	30	168	1264	4331	96	NA	16	633	353	2119	63	NA	81
Total TCDF	3199	1652	549	176	281	4444	2921	50943	86087	3721	NA	771	9767	17459	41390	5917	NA	6610
Total PeCDF	1980	915	287	40	98	2508	1759	36329	75322	1823	NA	354	6664	11597	32818	2993	NA	4567
Total HxCDF	847	516	126	21	38	1307	1167	25277	57162	931	NA	126	4323	6159	25014	1655	NA	1964
Total HpCDF	274	205	38	4	10	382	519	11214	28119	319	NA	34	2421	2540	11897	586	NA	614
Total OCDF	56	44	11	9	4	40	132	2396	7641	54	NA	6	710	536	2088	64	NA	74
TEQ	131	89	25	2	9	231	170	3398	6433	141	NA	27	581	887	2594	239	NA	346
PCDD/F Total	8999	3815	1305	257	467	9682	8986	144897	306758	8693	NA	1463	34369	48422	178480	12767	NA	15681
<b>Other Measured Parameters</b>																		
Average CO (ppm)	78	99	51	27	45	91	107	150	115	77	10	112	117	100	99	118	15	77
Average TC6 (°C)	168	121	170	120	131	132	136	150	172	117	10	102	96	157	129	124	5	133
Maximum CO (ppm)	222	320	175	82	117	186	178	337	299	159	13	182	401	278	221	215	18	211

NA - not applicable, no waste was consumed during blanks

ND - none detected

Table 3. Statistical Analysis<sup>a</sup> of Baseline Tests  
(PCDD/F concentrations in ng/kg waste consumed)

Parameter	Mean	Std. Deviation	Upper 95% Mean	Lower 95% Mean
2,3,7,8-TCDF	52.97	37.48	99.52	6.43
2,3,7,8-TCDD	4.57	1.93	6.97	2.18
1,2,3,7,8-PeCDD	9.52	6.01	16.98	2.07
1,2,3,4,7,8-HxCDD	7.11	4.33	12.48	1.73
1,2,3,6,7,8-HxCDD	10.52	7.85	20.26	0.78
1,2,3,7,8,9-HxCDD	21.92	16.94	42.95	0.88
1,2,3,4,6,7,8-HpCDD	42.23	34.57	85.16	-.069
1,2,3,4,6,7,8,9-OCDD	43.65	36.33	88.75	-1.46
1,2,3,7,8-PeCDF	43.21	33.34	84.62	1.81
2,3,4,7,8-PeCDF	73.01	52.49	138.17	7.83
1,2,3,4,7,8-HxCDF	121.82	94.53	239.19	4.44
1,2,3,6,7,8-HxCDF	43.12	33.73	85.00	1.25
2,3,4,6,7,8-HxCDF	67.78	64.12	147.40	-11.84
1,2,3,7,8,9-HxCDF	3.79	2.01	6.29	1.30
1,2,3,4,6,7,8-HpCDF	112.17	89.31	223.06	1.28
1,2,3,4,7,8,9-HpCDF	15.80	11.78	30.41	1.18
1,2,3,4,6,7,8,9-OCDF	33.61	24.46	63.98	3.24
Total TCDD	412.62	501.8	1035.61	-210.37
Total PeCDD	281.32	292.94	645.07	-82.41
Total HxCDD	221.49	234.22	512.31	-69.32
Total HpCDD	105.28	95.26	223.55	-13.00
Total OCDD	43.65	36.32	88.75	-1.46
Total TCDF	1880.42	1541.87	3794.88	-34.05
Total PeCDF	1020.46	860.84	2089.32	-48.40
Total HxCDF	491.76	405.91	995.76	-12.24
Total HpCDF	169.22	139.26	342.13	-3.69
Total OCDF	33.61	24.45	63.98	3.24
TEQ	79.15	60.30	154.02	4.29
Totals PCDD/F	4655.97	4020.02	9647.42	-335.49
Average CO (ppm)	82.72	39.06	102.13	63.29
Average TC6 (°C)	134.91	22.98	147.15	122.67
Max. CO (ppm)	200.77	103.65	252.32	149.23
Max. TC6 (°C)	478.30	92.81	527.75	428.85

a - measurements from all baseline runs were normally distributed